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PROVISIONAL APPLICATION FOR PATENT COVER SHEET

This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

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INVENTOR(S)						
Given Name (first and middle [[if any]) Family	Name or Suma	ne	(City and either	Residence r State or Foreign Country)	
William M.	Goldberger			Columbus, Oh	mbus, Ohio	
Mark S. Zak				Norwalk, CT	06850	
Additional inventors are being named on the separately numbered sheets attached hereto						
TITLE OF THE INVENTION (500 characters max)						
PROCESS FOR THE DIRECT REDUCTION OF IRON OXIDES IN AN ELECTROTHERMAL FLUIDIZED BED AND RESULTANT PRODUCT						
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ENCLOSED APPLICATION PARTS (check all that apply)						
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2 Drawing(s) Number of Sh	neets		Other (sne	clfu		
Application Data Sheet. See 37 CFR 1.76						
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The Commissioner is hereby authorized to charge filing fees or credit any overpayment to Deposit Account Number: 50/1039 \$80.00						
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United States Government.						
Yes, the name of the U.S Government agency and the Government contract number are:						
Respectfully submitted,	July B. Heller		Date	03/19/2002		
REGISTRATION NO. 30.181		30,181				
TYPED OF PRINTED NAME	epheri B. Heller	-	(If appropriate) Docket Number: GOLD 11			
(312) 236-8500 GOLD 119						

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PROCESS FOR THE DIRECT REDUCTION OF IRON OXIDES IN AN ELECTROTHERMAL FLUIDIZED BED AND RESULTANT PRODUCT

Inventors: William M. Goldberger Mark S. Zak

Background

reduction of iron oxides into metallic iron at temperatures below the melting point of iron (1530° C), commonly known as "direct reduction." More specifically, the present invention is directed to an improved process for the direct reduction ("DR") of iron ores when using solid carbon as the reductant in which the heat needed for the reduction reactions is provided by resistively heating the reactant materials while they are in a state of fluidization. The invention additionally provides a unique DRI product as a result of the inventive process.

[0002] Methods for the production of metallic iron from iron

minerals have evolved from times preceding the earliest recorded history. Iron ores of various types are found in enormous amounts throughout the world. Most iron ores are oxides of iron, mainly hematite (Fe_2O_3) . To make metallic iron, the ore is chemically reduced using agents that are in most cases derived from coal or natural gas (methane). The reduction of iron oxide to form metallic iron is endothermic, that is, heat is required to achieve the removal of oxygen atoms from the iron oxide molecule.

[0003] The method that has been the workhorse for the present industrial production of iron for steelmaking is the blast furnace. The furnace itself is a tall, very large diameter generally cylindrical steel vessel that is lined with refractories.

is fed with solid carbon (coke) into the top of the blast furnace. Air is introduced at near the base of the furnace to burn a portion of the carbon to generate carbon monoxide. This gas then reduces the iron oxide into metallic iron. The downward movement of the ore against the rising flow of reducing gas makes the blast furnace a highly efficient countercurrent reactor for the production of metallic iron from iron ores. The partial combustion of the coke provides the necessary heat for the reduction and for the melting of the metallic iron formed by the reduction. As the iron melts, it trickles downward through the unburned coke and forms a pool of molten metal which is periodically tapped and transferred to other

vessels where it is either solidified as a high carbon "pig iron" or treated in the molten state to remove carbon and to add alloying agents to form various steel products.

[0005] Despite the excellent thermodynamic and thermal efficiency of the blast furnace for making iron and steel, the recent trend in new plants for production of iron from iron ores is to use direct reduction (DR) processing. Indeed, the increase in iron making capacity in recent years has been largely by DR processing.

In there are a number of reasons for the rapidly increasing use of DR rather than the well established blast furnace method. For one, iron ore deposits often contain substantial amounts of very fine ore that is unsuitable for use directly in the blast furnace. A number of DR processes were originally developed to process iron ore fines without the need to first pelletize or agglomerate the ore.

[0007] Another important reason for choosing DR in place of the blast furnace is that the blast furnace requires "coking" coals. These coals, when heated to remove much of their volatile content, will form a coke material that has a lesser tendency to soften and provides the strength to support the weight of the burden of iron ore and coke that is moving downward in the furnace. Although there are numerous large coal deposits throughout the world, very few have the necessary characteristic of high strength after coking

to be useful in the blast furnace. As all nations desire to produce steel products internally rather than import them, those that do not have coking coals but have lower rank coals, or that have abundant natural gas, have built DR plants.

[0008] Another reason for the popularity of DR is the flexibility of producing a granular or briquetted iron product that can be shipped elsewhere for melting and production of steel, making the full investment for integrated steel an mill unnecessary.

[0009] DR processes are of two basic types depending on whether the reducing agent is gaseous or it is a form of solid carbon. Most operating DR plants use shaft furnaces using gaseous reductants derived from natural gas. These are known generally by their acronyms MIDREX and HYL. Although natural gas itself is not an effective reductant, it can be converted into hydrogen or mixtures of hydrogen and carbon monoxide by "reforming". The reforming of natural gas is well established technology that involves several process reactors and catalytic conversion, thus entailing significant capital and operating costs.

[0010] Gaseous reduction systems are also generally operated at elevated pressure to increase the productivity per unit of reactor volume so that the vessel size can be held within the limits of practical construction. The need to operate at elevated pressure requires expensive pressure vessels and the solid feed and product

handling must be done using "lock hoppers." This adds significantly to the cost of the DR plant.

[0011] Gaseous reduction has another disadvantage, the reduction reactions are slow at low temperature, and it is necessary to operate the reduction reactors at above 650°C to achieve reasonable processing rates. The direct partial combustion of the reducing gases within the DR reactor is a possible means to achieve the heating, but this procedure is difficult to control and can be dangerous. Instead, the ore is preheated in a separate reactor to a temperature above the desired reduction temperature so that the excess heat provides the heat needed to maintain the desired temperature for reduction. Additionally, the reducing gas is generally preheated by indirect heat exchange before introducing it into the reduction reactor. Preheaters add complexity and cost and are not a highly efficient means to add heat to the reaction system.

[0012] For locations that do not have natural gas, DR processes have been developed that use coal as a solid reductant. The predominant processes of this type have used long, horizontal refractory lined rotary kilns as the process vessel. These processes are known generally by their acronyms SL/RN, DRC, and ACCAR/OSIL. Coal, iron ore and some limestone are fed to the kiln which is operated at temperatures between about 850-1,050°C. Auxiliary burners fired with various fuels, including pulverized

coal, are generally provided to heat the charge materials from above. The rate of the reduction reactions with relatively coarse solid carbon and iron ore is not rapid, and the plants using this type of reducing reactor are limited in capacity to generally less than 100,000 metric tons per year. The rotary kiln processes are subject to the problem of the tendency for the formation of accretions on the wall of the kiln. The productivity also varies with the type coal. Although rotary kiln based DR plants have been built in many places of the world, the combined annual capacity of these is less than 2.5 million metric tons, which constitutes less than 5 percent of the total DR production capacity.

employs a rotary hearth rather than a rotary kiln. These processes are known generally by their acronyms Inmetco and FASTMET. Several rotary hearth plants have been built, but, like the rotary kiln plants, these have also been of limited capacity. To improve the reaction kinetics and to avoid the tendency for agglomeration to occur within the reducing reactor, the rotary hearth processes have used a feed of pelletized iron ore fines which also contain the carbon reductant within the pellets. Fine coal is generally used as the reductant. The intimate contact between the fine particles of iron ore and the fine carbon provides a relatively rapid rate of reduction compared with the rotary kiln processes. The heating is also done through use of auxiliary burners positioned in the roof

of the rotary hearth furnace. These can be operated with various Heat may also be generated by burning a portion of the combustible gases produced by the reduction reactions and any volatile matter that evolves from the coal. The source of heat within the rotary hearth furnace, as in the rotary kiln, generated above the charge material, and the rate of heating is largely by radiation from the flames above. To achieve reasonable rates of heat transfer to the charge to sustain the reduction, it is necessary to limit the thickness of the layers of pellets on the hearth to only two or three layers. Higher temperatures (1,250-1,450°C) are used than in the rotary kiln processes and, with the use of pellets containing the reductant, the reduction rate is higher. However, the throughput is limited by the restriction of having only very thin layers of the reacting mixture on the hearth. [0014] It is worth noting that while most of the world capacity for directly reduced iron (DRI) is based on processes that employ gaseous reductants in moving-beds (shaft kilns), such as the MIDREX and HYL processes, the more recently built DR plants have selected fluidized-bed processes, known by their acronyms FIOR/FINMET and As discussed above, the fluidized-bed processes offer CIRCORED. the advantage of processing iron ore fines without requiring pelletization. They also offer very efficient contact between the particles of iron ore and the gaseous reductant. In addition, they provide extremely high heat transfer rates and

temperature control. However, the fluidized-bed processes of the prior art have all used gaseous reductants. No prior art DR process has used fluidized-bed technology for the direct reduction of iron ore with solid reductants.

[0015] Accordingly, it is the primary object of the present invention to provide a process for the direct reduction of iron oxides in a fluidized-bed reaction vessel using solid carbon as the reducing agent.

[0016] It is also an object of the present invention to provide a process for the reduction of iron oxides in a fluidized-bed at significantly higher temperatures, and therefore higher reduction rates, than that achieved in the present processes for the fluidized-bed gaseous reduction of iron oxides.

[0017] It is a further object of this invention to provide the heat needed to sustain the reduction reaction and maintain the desired reaction temperature by use of highly thermally efficient electrothermal or charge-resistance heating.

[0018] It is still another object of this invention to increase the production rates to rates that are significantly higher than those reported for the prior art DR processes that employ solid reductants.

[10019] Other objects and advantages of the process of this invention will become evident from the following description of the process, equipment and methods of operation.

Brief Description of the Drawings

[0020] The foregoing and other objects will be more readily apparent by referring to the following detailed description and the appended drawings in which:

[0021] Fig. 1 is a vertical cross-sectional view of an electrothermal fluidized bed furnace for use in the present invention.

[0022] Fig. 2 is a schematic diagram of the process for an improved method of achieving rapid and efficient reduction of iron oxide in an electrothermal fluidized bed furnace.

Detailed Description of the Preferred Embodiment

ore pellets is conducted in a fluidized bed comprising a mixture of coarse carbon particles (pure synthetic graphite, coke, etc.) and partially-reduced iron ore pellets. Carbon particles provide electrical conductivity for the fluidized bed and heat generation internally within the fluidized bed is due to passing the electrical current through fluidized bed. Further, the carbon-rich fluidized bed zone helps to reduce the product gases back from CO₂ to CO and, if water is present, to CO and H₂. The dimensions of the dry green pellets and carbon particles are chosen to provide uniform fluidization of both materials with a degree of reduction of less than 25-35%. The apparent specific density of DRI pellets

changes from 2.1-2.25 g/cc, for pellets with Fe_2O_3 content of approximately 75% (and approximately 50% of total iron), to 4.2-5.2 g/cc for reduced pellets with total iron content of approximately 80-85% and Fe_2O_3 of approximately 5-7%. The shrinkage in volume of DRI pellets is between approximately 25-35%. It is believed that the physical characteristics of the reduced pellets may also differ from the reduced pellets from other direct reduction processes.

It has been found that essentially complete reduction of the iron ore contained within the pellets occurs with less than the stoichiometric consumption of the carbon contained within the pellets. This suggests that some of the free coke in the fluidized bed serves as a reductant as well as providing electrical conductivity. the carbon added to the green pellets Thus conceivably could be reduced or possibly eliminated. This also suggests that the pelletizing steps may be eliminated, and that iron ore that is crushed and sized could be directly introduced into the electrothermal fluidized bed ("EFB") furnace after preheating. The amount of carbon in the reduced pellets can also be adjusted to suit the needs of the subsequent steel production. Turning to Fig. 1, a mixture of "green" iron ore pellets containing carbon as the reductant and having a bulk density of about 2.1-2.5 g/cc is mixed with granular carbon having a bulk density of 0.9-1.1 g/cc and fed into the EFB furnace 50 through inlet 60 and falls by gravity through freeboard space 62 above

upper section 52 and onto the upper part of the fluidized-bed of material contained in section 52. Fluidizing gas is made to enter the base of EFB furnace 50 and into lower section 54 through nozzles 66. As the pellets become reduced, the density of the pellets increases to a density higher than that which can be maintained suspended by the rising gases within section 52. These denser pellets have a tendency to move downward into the lower conical or tapered section 54 where the gas velocity is higher because of the smaller cross-sectional area and thus is capable of fluidizing the denser partially reduced pellets. Because the coke particles are substantially less dense than either the green pellets or the partially reduced pellets, the coke tends to remain in the upper section 52, whereas the denser, partially reduced pellets are contained mainly in lower section 54. However, because of the substantial turbulence created by the high velocity of the gas entering section 54, there is considerable mixing of the material contained within the two zones of fluidization. When the iron ore pellets are fully reduced, the density becomes 4.2-5.2 g/cc, and at this higher density they move to the lower part of conical section 54 and can be removed through discharge device 64. The electrothermal heating of the furnace is accomplished [0026] by locating one or more vertically-oriented electrodes of a first polarity spaced from the walls of upper section 52. One or more electrodes 58 of opposite polarity are located along the inner

walls of upper section 52. A voltage is then applied to cause electric current to flow from electrode(s) 58 through the fluidized-bed of material contained in section 52. The presence of substantial carbon in section 52 provides sufficient electrical conductivity to allow current to flow and generate the heat needed for the reduction by direct resistive heating of the material in section 52. Although the resistive heating occurs essentially entirely within section 52, the high degree of mixing of the materials in section 54 and section 52 provides effective heating of the denser iron ore particles throughout the time of their residence within the EFB furnace such that the reduction reactions continue to completion.

within the furnace is dependent on the amount of material contained within the furnace such that the greater the amount of material contained, within the furnace such that the greater the amount of material contained, the longer will be the retention time for same rate of feeding of the pellets to the furnace. It follows therefore that for a furnace of a given cross-section, the residence time can be varied by varying the feed rate and by independently varying the height of the material in section 52 to accommodate a greater amount of material. This feature illustrates a major difference in the fluidized-bed furnace from the rotary hearth type that has limited ability to increase residence time except by reducing the throughput.

Referring now to the flow sheet of the inventive method and apparatus for reduction of iron oxide in an EFB furnace shown in Fig. 2, the iron ore is first pelletized in the well-known manner. To this end, the process includes three feed bins, 10, 12 and 14, that contain the raw materials for the process. Feed bin 10 contains iron oxide (iron ore fines, iron oxide concentrates, etc.); feed bin 12 contains solid reductant (green petroleum coke, pulverized coal, coke breeze, etc.); and feed bin 14 contains binder (bentonite, organic resin, etc.).

[0029] Raw materials from the feed bins 10, 12 and 14 are mixed together in proper proportions in a mixer or blender 16. Preferably, the dry mixture contains approximately 75 percent (by weight) iron oxide, up to approximately 25 percent (by weight) solid reductant, and approximately 1 to 2 percent (by weight) binder. The mixture 17 is then processed through a size reduction mill 18 to reduce the components to a generally uniformly-sized powder. The powdered materials are then introduced to a pelletizer 20, where water is also added, to form "wet" green pellets 21.

[0030] The green pellets 21 are transported to a dryer 22 where they are dried at between approximately 110-130°C (230-260°F) to remove moisture to less than a residue moisture content 0.5-1.0%. The exhaust gases from the dryer 22 are passed through a cyclone 23. Any dust in the gas is recycled back to the mixer 16 for reprocessing into green pellets. The exhaust gas is then passed

through an electrostatic precipitator 24 before being exhausted to atmosphere.

[0031] The dry green pellets 25 are then divided by screener 26 to separate out pellets that are sized between 6 mesh and 40 mesh (-3.5 + 0.425 mm). The pellets larger than 6 mesh are sent to a size reduction mill 27 to be reduced in size, and then returned to the screener 26 to be redivided. Pellets smaller than 40 mesh are recycled directly to the pelletizer 20 to be reformed.

The dry green pellets with sizes between 6 and 40 mesh [0032] are loaded into the bin 29, and from it into an EFB furnace 32. As illustrated, the dry pellets are passed through a heater 34 before entry into the EFB furnace 32. Free fresh coke (coal, coke or other carbon-containing particles not within the pellets used to provide electrical conductivity and for fluidized stabilization), which may be supplemented with coke recovered as the non-magnetic fraction by magnet separation of the mixture of coke and reduced pellets, is loaded into the EFB furnace from the bin 30 in proper proportion with the dry green iron oxide pellets, preferably in a ratio of between about 3:1 and 5:1 (by weight) of iron oxide pellets to free coke. Any low cost, high content carbon material may be used. In experimental use, a high purity carbon additive commercially available from the Superior Graphite Co. of Chicago, Illinois, U.S.A. as Desulco® 9010, has been used.

[0033] The solid particles are fluidized by a gas, which may be

nitrogen and/or recirculated furnace gases, introduced into the bottom of the conical section of EFB furnace 32. The green pellets are reduced in the fluidized bed at temperatures between approximately 900-1,080°C (1,652-1,980°F) and residence times of between approximately 15-75 minutes. The reduced DRI pellets 37 and minor amounts of free coke or individual carbon particles 38 are discharged from EFB furnace 32, cooled in a heat exchanger 36, and sent to a separator 39. The reduced DRI pellets with a total iron content of 85-95% and a carbon content of 5-15% are separated from the free coke by common physical separation methods, such as magnetic separation.

[0034] After separation from the reduced pellets, the free carbon is returned to the bin 30 and loaded back into the EFB furnace 32 for fluidized bed stabilization. The flue gas 40 from the EFB furnace may be cleaned in a cyclone 42. The collected dust is sent back to the bin 30. If a large amount of iron ore is in the dust, the dust can be stockpiled and the fine particles of iron ore separated out. The flue gas from the cyclone 42 is cleaned by a bag house or electrostatic precipitator 44 and may be either burned and exhausted to the atmosphere or recycled. As shown, part of the clean mixture of nitrogen and carbon monoxide 43 is recycled and burned for pellet drying in the dryer 22, while part of the EFB furnace flue gas 40 can be used directly, without cleaning, for preheating of the pelletized iron ore in the heater 34 before

introduction into the EFB furnace 32. The hot exhaust from the heater 34 can also be recycled to the pellet dryer 22.

[0035] As discussed above, an EFB furnace is provided that is particularly suited for use in the direct reduction of iron oxide set forth above. With reference to Fig. 1, the EFB furnace, generally designated 50, comprises a housing having an upper fluidized bed section 52 with vertical walls and a generally constant cross-sectional area and a lower fluidizing gas distribution section 54 with sloping walls and a cross-sectional area that decreases from its upper end toward its bottom.

[0036] As seen in profile, the lower portion 54 has a conical appearance. Importantly, the slope of the walls in the lower section must be steeper than the angle of repose for the reduced pellets so that none collect on the walls of the lower section. The reduced cross-sectional area of the lower section 54 promotes the separation and segregation of the free coke from the pellets and the re-circulation of the pellets into the upper section 52 of the furnace 50.

[0037] The upper section 52 of the furnace includes one or more vertically-oriented electrodes 56 of a first polarity that are spaced from the walls of the upper section 52. One or more electrodes 58 of the opposite polarity are associated with the walls of the upper section 52. In operation, the current that passes between the electrodes 56 and 58 provides the heat for the

furnace 50.

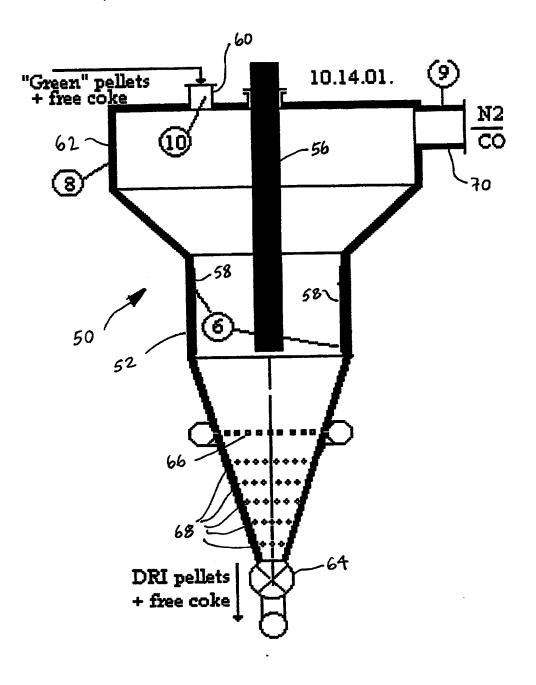
[0038] Green pellets and free coke are fed into the EFB furnace 50 through an inlet 60 in a freeboard space 62 above the upper section 52. As the pellets are reduced, the density of the pellets increases, and a point will come when the fluidizing gas can no longer suspend the pellets. Then, the DRI pellets, and any free coke associated therewith, exit the EFB furnace through a discharge feeder 64 at the bottom of the lower section 54.

Fluidizing gas is introduced into the lower section 54 [0039] through a series of inlet nozzles 66, which are spaced about the lower section to provide an even distribution of the gas. Optionally, in the illustrated embodiment the fluidizing gas is fed to the nozzles through a tubular heat exchanger 68 that encircles the lower section 54 before connecting to a manifold that includes In this way, the temperature of the the inlet nozzles 66. fluidizing gas is raised before it is injected into the furnace, while the temperature of the reduced pellets that pass through the lower section 54 is lowered to a temperature more likely to inhibit reoxidation and agglomeration of the reduced pellets. Optionally, a separate heat exchanger may be provided below the discharge feeder 64 for which the fluidizing gas may also be used as the coolant (as shown in Fig. 1). The spent fluidizing gas and the gaseous byproducts of reduction (CO) exit the freeboard space 62 through a flue outlet 70. The gas can be used advantageously used as fuel in related steel making steps in an integrated steelmaking facility to, e.g., melt the reduced pellets, resulting in potential cost savings from reducing the need for other energy sources.

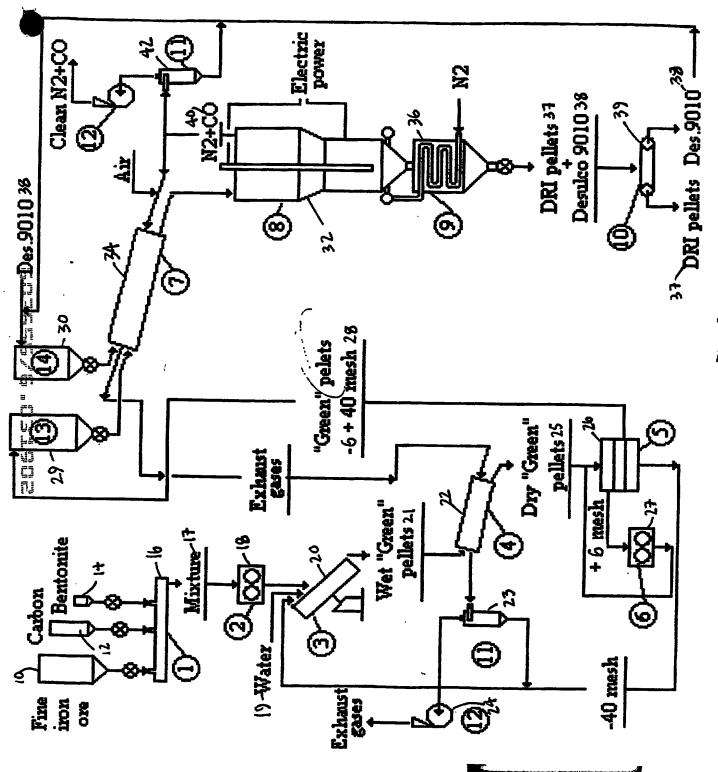
[0040] Accordingly, a DR process meeting all the objects of the present invention has been provided. The EFB furnace provides for a controlled thermal reaction and the reduced cross-section of the fluidizing gas distribution portion of the furnace promotes segregation of the free carbon from the reduced pellets. results with this process show that properly formed dry pellets mixed with free coke with a weight relation from approximately 3:1 to approximately 5:1 can be reduced in EFB furnace with the fluidized bed formed initially by the same type of coke at temperatures between approximately 900-1,080°C (1,652-1,980°F) with direct furnace heating by electrical power without agglomeration of reduced pellets. The reduction of iron oxide pellets with small sizes and low inside resistance of heat transfer and diffusion increases the specific process productivity by 2-4 times at temperatures 250-400°C less than processes using hard carbon The use of electrical power for direct heating reductants. provides for a simple furnace design. In addition, efficiency can be improved by burning of carbon monoxide and other volatiles produced in process of pellet reduction for green pellet drying and DRI pellet melting.

ABSTRACT

A method and an apparatus for producing direct reduced iron from dry pellets composed of iron oxide and carbonaceous material. A mixture of pellets and free coke particles with weight relation from 3:1 to 5:1 is fed into the top of an electrothermal fluidized bed that is fluidized by nitrogen. By exposing pellets in an electrothermally-heated fluidized bed to temperatures of between approximately 900-1,080°C for an average period of between approximately 15-75 minutes, the volatiles are removed and the pellets metallized. Reduced pellets mixed with free coke are discharged from the bottom of fluidized bed and cooled. The reduced iron pellets are physically separated from any free coke, and the free coke is recycled back into the fluidized bed.



F1G. 1



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F16, 2